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Measurement of neutron activated short-lived nuclides using a pneumatic transfer system

Paas, Alfred O.; Sullivan, Robert D.

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MEASUREMENT OF NEUTRON ACTIVATED SHORT-LIVED NUCLIDES USING A PNEUMATIC TRANSFER SYSTEM

* * * * *

Alfred O. Paas

and

Robert D. Sullivan

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by

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and

Robert D. Sullivan
Captain, United States Army

Submitted in partial fulfillment of
the requirements for the degree of

MASTER OF SCIENCE
IN
PHYSICS

United States Naval Postgraduate School
Monterey, California

1962

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This work is accepted as fulfilling

the thesis requirements for the degree of

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IN

PHYSICS

from the

United States Naval Postgraduate School

D. W. Rodeback
Faculty Advisor

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Chairman
Dept. of Physics

Approved:

B. R. Linvell
Academic Dean

ABSTRACT

A rapid pneumatic transfer device has been designed and constructed for use with the U.S. Naval Postgraduate School AGN-201 Reactor. This device permits rapid removal of the sample from the center of the core of the reactor to a shielded counting position to permit measurement of the half-lives of short-lived nuclides. Preliminary measurements of the decay of a mixture of Hf^{178m} and Hf^{179m} were made. The Hf^{178m} and Hf^{179m} nuclides were produced by the neutron activation of a Hafnium sample. Preliminary measurements of the resulting decay were made by detection of the gamma emission. Decay data were obtained with the use of a scintillation counter and a multi-channel analyzer. The measured half-life of Hf^{179m} was 18.2 seconds compared to the published value of 19 seconds. The half-life of Hf^{178m} has not been determined because of limitations of the analyzer. Proposed methods to improve measurements of both of these half-lives are discussed. Computer programs were devised for least squares fitting of the decay of an independent mixture of two nuclides and for single nuclide decay. These programs are described, and the FORTRAN statement for the decay of a mixture of two nuclides is included.

The authors wish to express their appreciation to Professor G.W. Rodebacl for his assistance and encouragement during the course of this undertaking. Appreciation is also extended to Professors W.W. Hawes, E.A. Milne, C.D.G. King, H.E. Handler, and D.G. Williams, reactor operator F.M. Spreckhardt, and to the many others who assisted in many phases of the project.

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I. OBJECTIVE

The objectives of this thesis were the following:

1. To design and construct a rapid pneumatic tube to be used in the AGN-201* reactor to permit the rapid removal of an irradiated sample from the core region of the reactor to a shielded counting position.
2. To utilize this pneumatic tube to measure half-lives of short-lived nuclides activated by neutron capture.

*The AGN-201 is an experimental thermal reactor with a core which is a homogeneous mixture of 20% enriched Uranium Dioxide and Polyethelene. The Reactor is graphite reflected and has a maximum flux of approximately 5×10^{10} neut/cm^2-sec at the maximum authorized power of 1 K.W.

II. TRANSFER SYSTEM

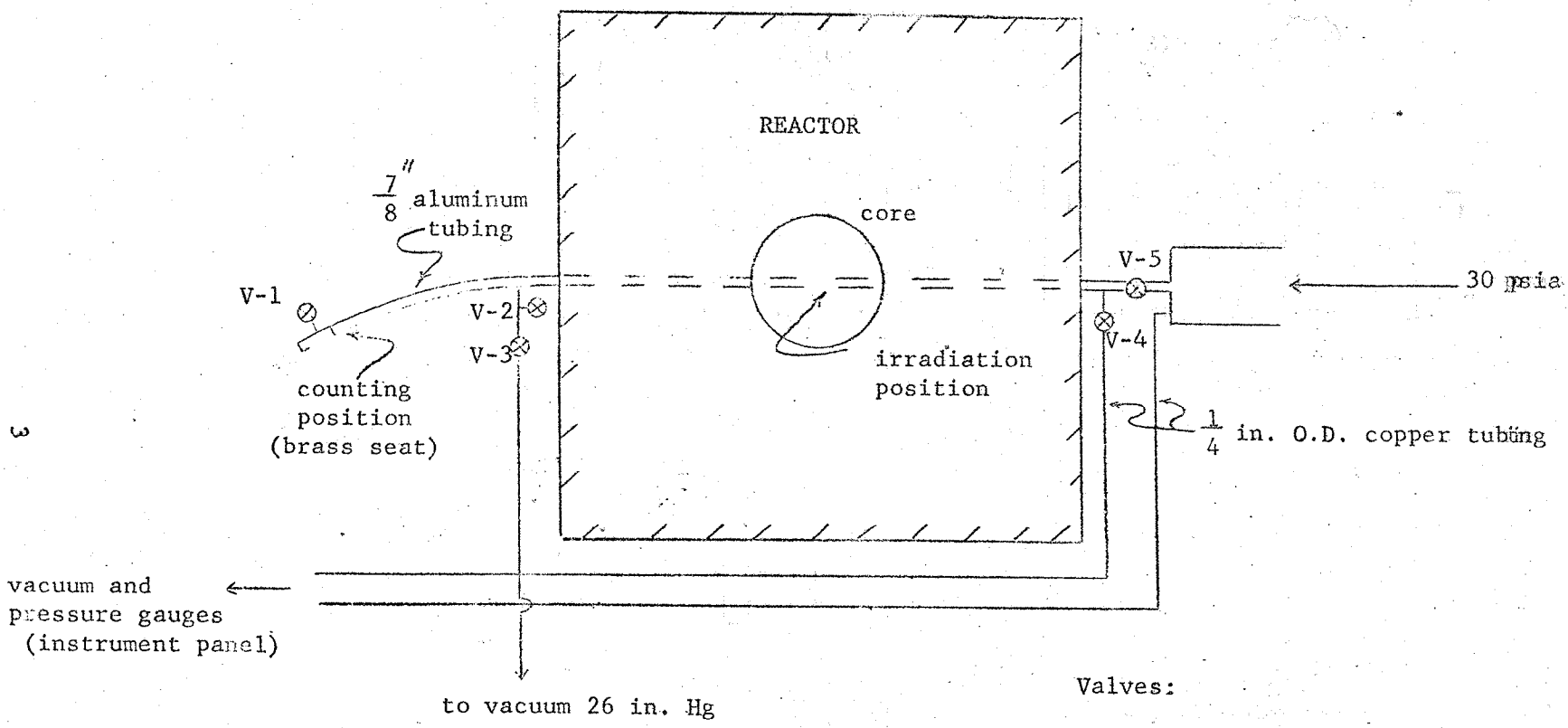
A. General

In order to determine half-lives which are in the order of seconds a procedure must be used that allows the decaying nuclide to be counted almost immediately after its removal from the source of activation. For our measurements this was accomplished by the use of a rapid pneumatic tube. The tube contained a sample holder or "rabbit" which was alternately transfered from an activation position, which was the core center of the reactor (maximum flux region), to a shielded counting position approximately six feet outside of the reactor (Figure 1). The movement of the rabbit from activation to counting position was accomplished by the release of high pressure air thru a quick-opening solenoid valve. The time of travel was approximately one-tenth of a second. When necessary the rabbit was returned to the core region for further activation of the enclosed sample by evacuating the tube. This brought the rabbit back onto its' seat at the activation position. The cycle could be repeated many times.

B. Pneumatic Tube and Accessories

The pneumatic tube was constructed from 7/8" outside diameter aluminum alloy tubing having a wall thickness of .049" (Federal Specification WW-T-778b). Since maximum available length of this tubing was twelve feet and a twenty-foot length was needed to extend through the reactor, two pieces were joined by welding, using an internal sleeve, which also served as a seat for the rabbit. The tube was inserted through the glory hole of the reactor and the rabbit was positioned by the seat at the point of maximum flux at the center of the core.

Auxiliary equipment attached to the aluminum tube consisted of five normally-closed solenoid valves which controlled pressure and vacuum in the tube, and a brass stopping seat. The seat is shown schematically in Figure 2 and pictured in Figure 3. By cycling the valves, the rabbit was moved from irradiation position to counting position and back again. Cycling was accomplished manually with a set of five switches located on the main instrument panel in the reactor room. This panel also contained pressure and vacuum gauges for the system.



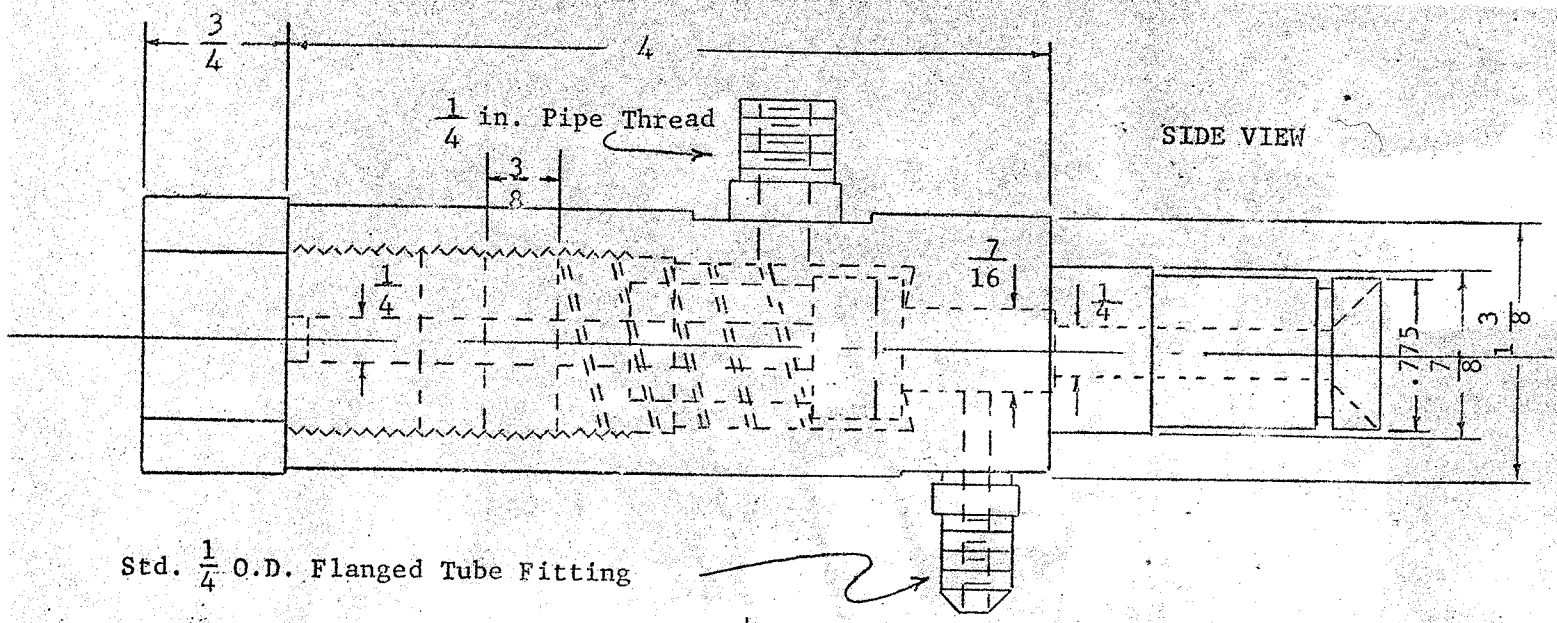
Valves:

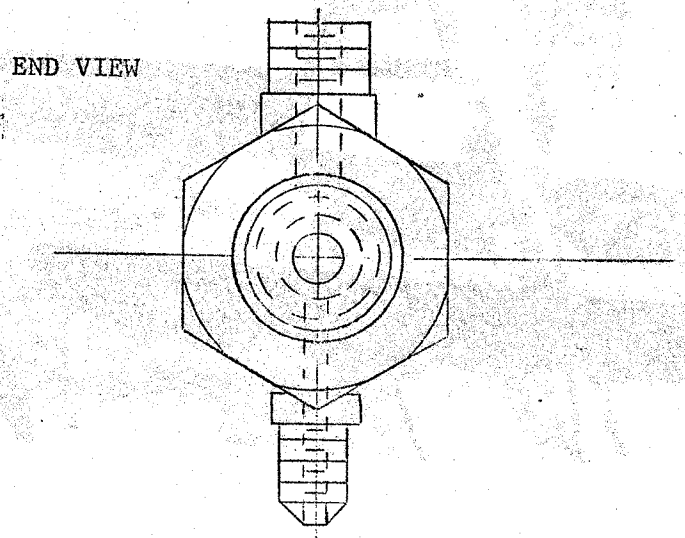
V-1, V-2, V-3, V-4 Skinner miniature two way solenoid valves, C series No. C2DA1092

V-5 Skinner two way high flow solenoid valve, L series No. L2DB5150

PNEUMATIC TUBE (Top View)

Figure 1. Pneumatic Tube





SCALE: FULL

All dimensions are in inches

FAST PNEUMATIC TUBE
ONE WAY VALVE and SEAT ASSEMBLY

Figure 2. One Way Valve and Seat Assembly

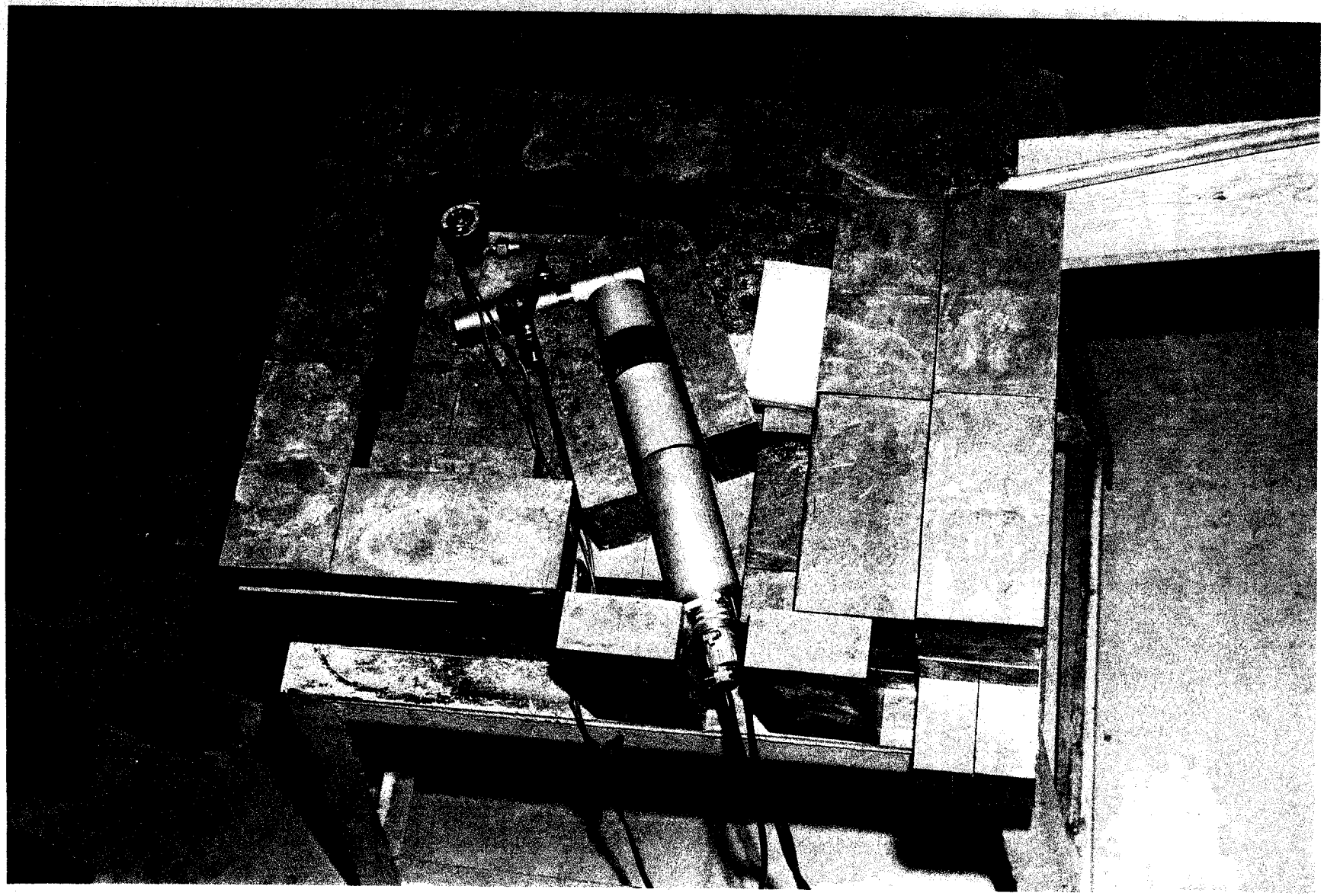


Figure 3. One-Way Valve and Detector (Top View)

With reference to Figure 1 a complete cycle of the rabbit is described. The rabbit is placed into the tube manually at the counting position by temporarily removing the brass seat assembly at the forward end of the tube. Valve V-4 is opened. This places a vacuum throughout the entire rear portion of the tube since valve V-3 is closed. The rabbit will move somewhat but will not enter the reactor. This is because of an equilization of pressure in the forward portion of the tube after the rabbit starts moving towards the core. To place the rabbit in the irradiation position (core) at a pre-set time, valve V-1 is opened to the atmosphere. This creates a pressure differential of approximately one atmosphere in the forward portion of the tube and sends the rabbit back onto its seat. Valve V-1 is then closed and valve V-3 is opened in order to place a vacuum in the entire tube. After irradiation of the sample (which is contained in the rabbit) it is now desired to move the rabbit to the counting position. First valves V-3 and V-4 are closed and then valve V-5 is opened, releasing an air pressure of thirty pounds per square inch against the rear of the rabbit. This sends the rabbit very rapidly forward and into the counting position. With the rabbit in this position, valve V-2 is opened for a short time to reduce the pressure in the tube to atmospheric. The cycle is then ready to be repeated when necessary.

An accessory piece of equipment used in this design was a 1.5 cubic foot welded steel air pressure tank tested to 120 psi (Figure 4). This was connected to an existing air compressor housed outside of the reactor building. A pressure reducing valve and an air filter were installed in the 1/2 inch line between the compressor and this tank. One end of valve V-5 (quick opening) was connected directly onto the tank. The opposite end was fitted over the rear end of the aluminum tube by means of an '0' ring connection held with three set screws. The vacuum system consisted of a Centro-Pressovac 4 Pump driven by a 1/4 horsepower motor connecting a vacuum reservoir of 314 cubic inch volume. All lines servicing pressure and vacuum to the aluminum tube through the solenoid valves were 1/4 inch outside-diameter, double strength copper tubing. Standard brass fittings were used at all connections.

Electrical connections between the solenoid valves and the individual switches on the instrument panel were made by means of 115-volt, two-strand, copper wire.

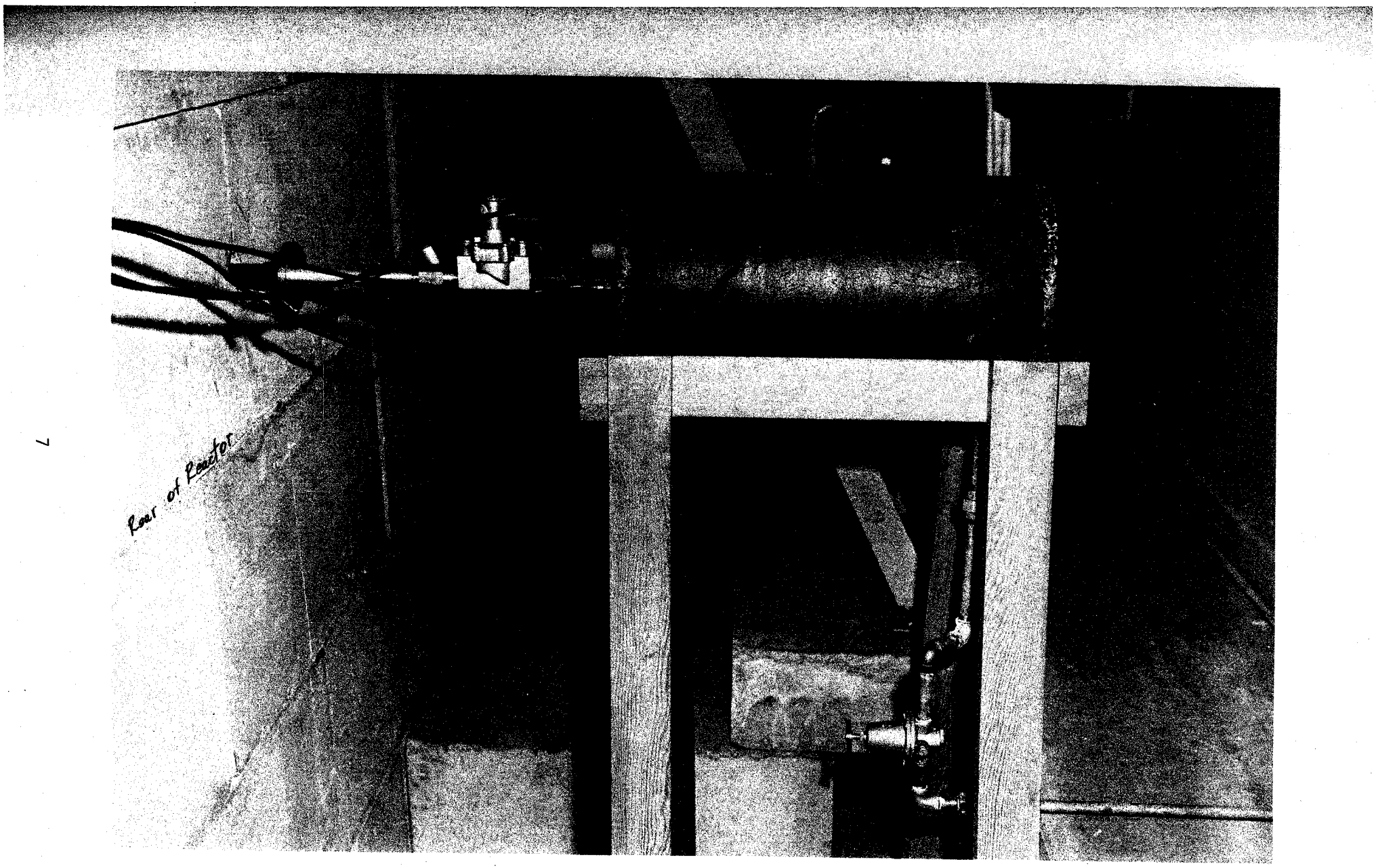
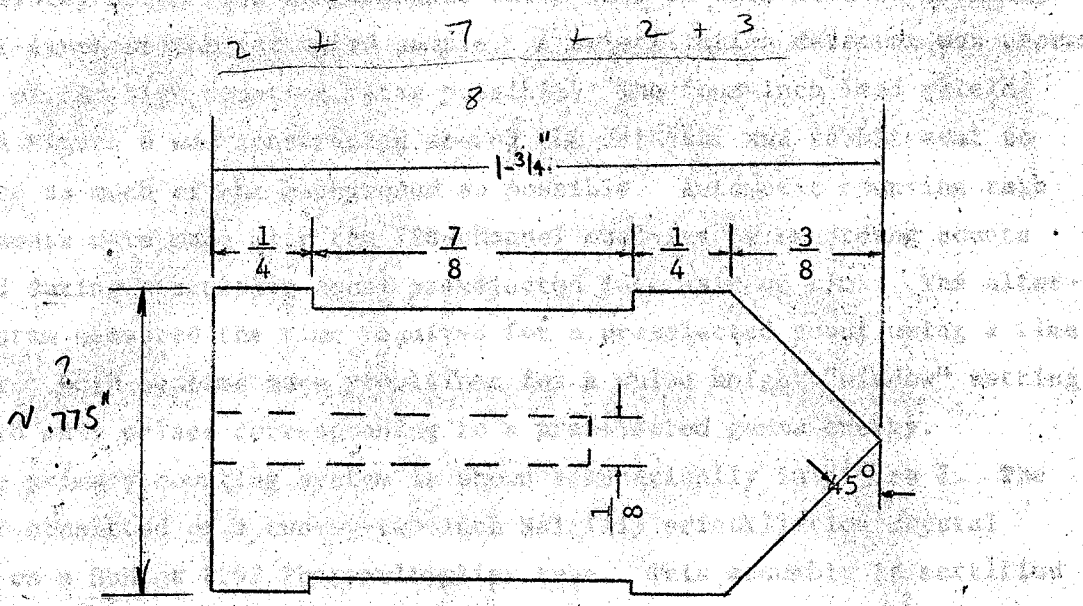


Figure 4. Pressure Tank (Side View)

There were two types of seating arrangements for the rabbit in the tube. As previously mentioned, the rabbit was stopped in the irradiation position by means of a sleeve inserted in the aluminum tubing at the desired place. This also served to strengthen the welded joint. However the seat (or stop) that was located in the counting position (Figure 2), contained an adjustable spring-loaded valve which permitted only one-way flow of air from the tube to the outside. The operating condition of this valve was set by adjusting the tension of the enclosed spring with a threaded holding nut. This spring in turn held a rubber gasket over the discharge opening of the valve. The valve was capable of passing air through the opening at a discrete rate determined by the tightness of the holding nut and by the pressure differential between the air in the tube and the atmosphere. The necessity for this type of valve was the slight build-up of air pressure in front of the rabbit, (since there was not a complete vacuum in the tube prior to firing). This build up can be gated through the seat valve at such a rate as to allow some of the air to slow the rabbit down and prevent it from striking the seat with excessive force. This slowing down takes place in the last foot of travel and can be adjusted as necessary to give variable slow-down times.

C. Rabbit

The rabbit was constructed from balsa wood as shown in Figure 5. It weighed 1.07 grams including the hafnium sample. The latter was in powdered form and was contained in a gelatin medicinal capsule within the 1/8 inch diameter hole located along the long axis of the rabbit. Securing of the rabbit was accomplished by a 1/8 inch plug of balsa wood inserted and glued in the outer portion of the hole. Various other holding positions for the sample can be devised dependent upon the physical properties of the sample to be activated.



RABBIT

All dimensions are in inches

Figure 5. Rabbit

III. COUNTING APPARATUS

After the neutron activated sample was removed from the reactor core and seated in the counting position, the counting apparatus was used to make disintegration rate measurements which were in turn used to determine the half-lives of the activated sample. A scintillation detector was chosen because of the high counting rates possible. The four-inch lead shield shown in Figure 6 was constructed around the detector and rabbit seat to eliminate as much of the background as possible. Automatic counting rate measurements were made with the 128-channel analyzer by recording counts observed during successive equal preselected intervals of time. The alternate system measured the time required for a preselected count using a time recorder. Both systems have provisions for a pulse height "window" setting to record only pulses corresponding to a preselected gamma energy.

The primary counting system is shown schematically in Figure 7. The detector consisted of a two-by-two-inch NaI (Tl) scintillation crystal mounted on a DuMont 6292 Photomultiplier tube. This assembly is certified by the manufacturer to be capable of 7.6% resolution (defined as the energy width at half maximum of the photopeak divided by mean photopeak energy) of the .662 MEV Cs^{137} gamma. The scintillation counter was used with an external high voltage power supply to replace the internal high voltage power supply (in the RCLiac 128-channel analyzer) which was found to have considerable drift. For measurement of counting rates with this system the "gross mode" of analyzer operation was used. The number of counts observed during each of 125-constant preselected time intervals were photographically recorded from the visual display of the analyzer. The counts corresponded to a given gamma energy range (determined by the window setting). The proper window setting was readily determined by using the 128-channel "pulse height analyzer mode" of operation. (See calibration, page 16).

An alternate counting system is shown schematically in Figure 8. It includes the scintillation crystal described above, a cathode follower, and a non-overloading linear amplifier with provisions for pulse height window settings. The decimal scaler has been modified to deliver pulses at 100 or 1000 count intervals to a two channel recorder. The second channel of the recorder was used for recording time calibration pulses. The high voltage for photomultiplier operation was supplied by a regulated high voltage power suppy. The occurrence of each 100 or 1000 counts was recorded as a

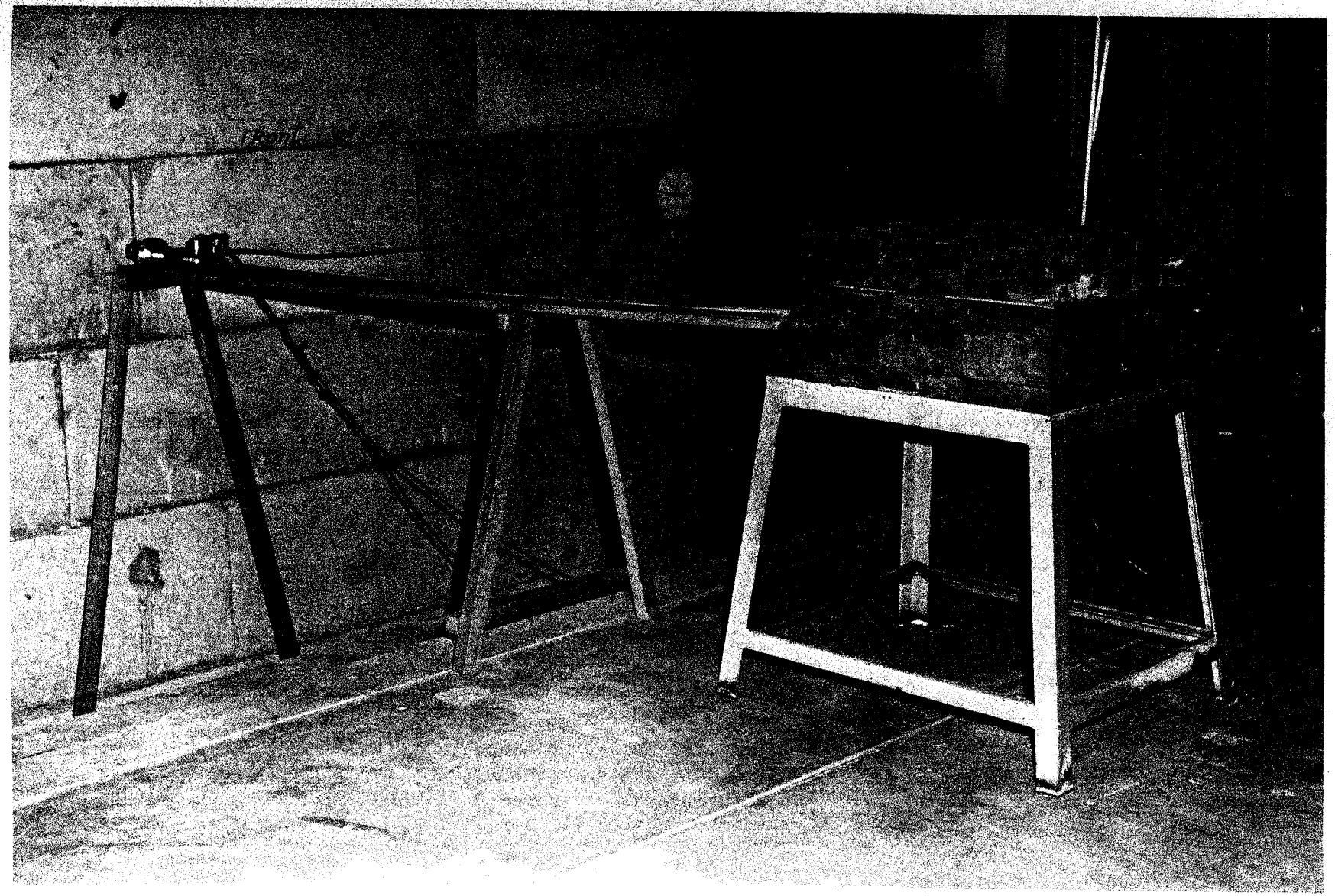


Figure 6. Lead Shield

COUNTING SYSTEM USING 128 CHANNEL ANALYZER

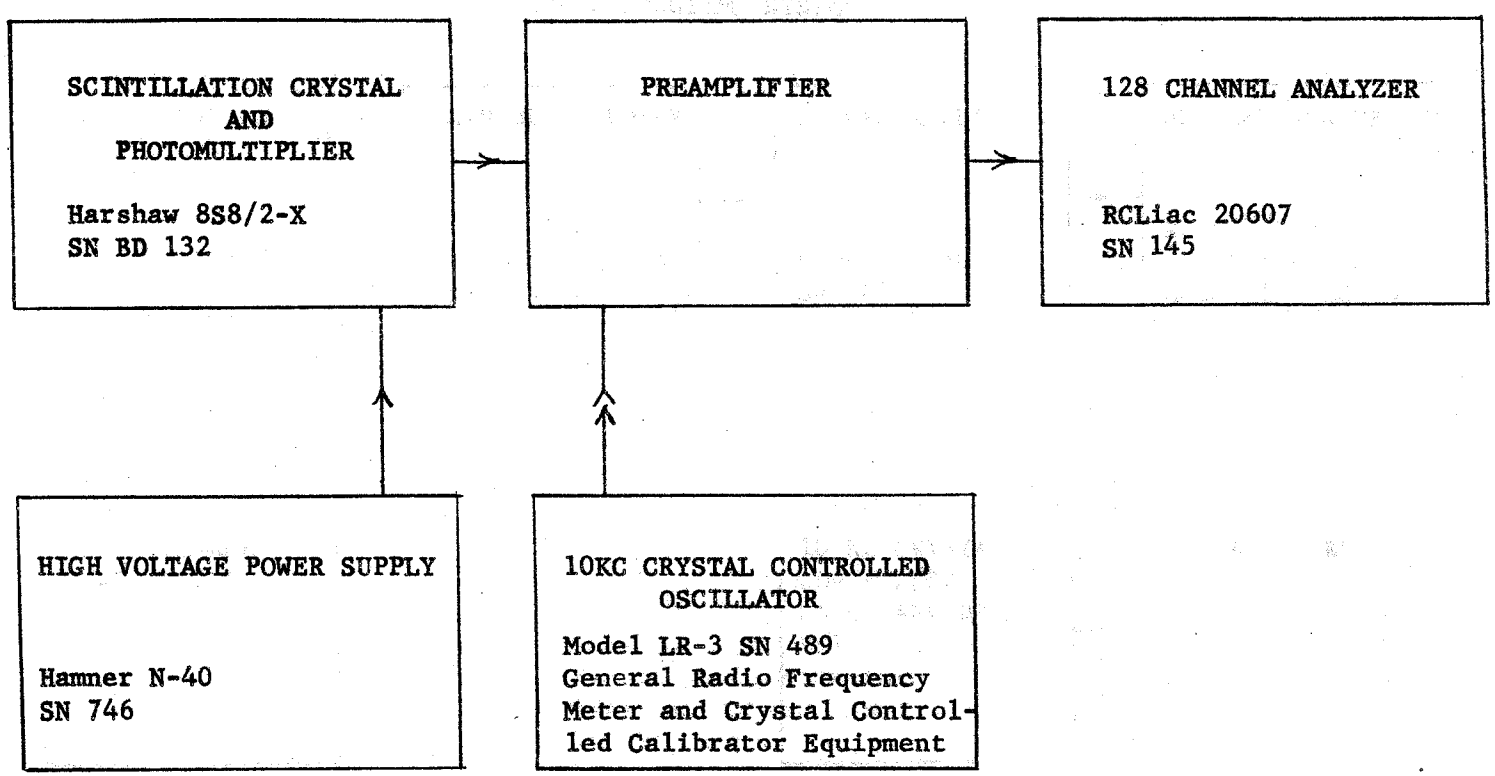


Figure 7. Counting System Using 128-Channel Analyzer

ALTERNATE COUNTING SYSTEM

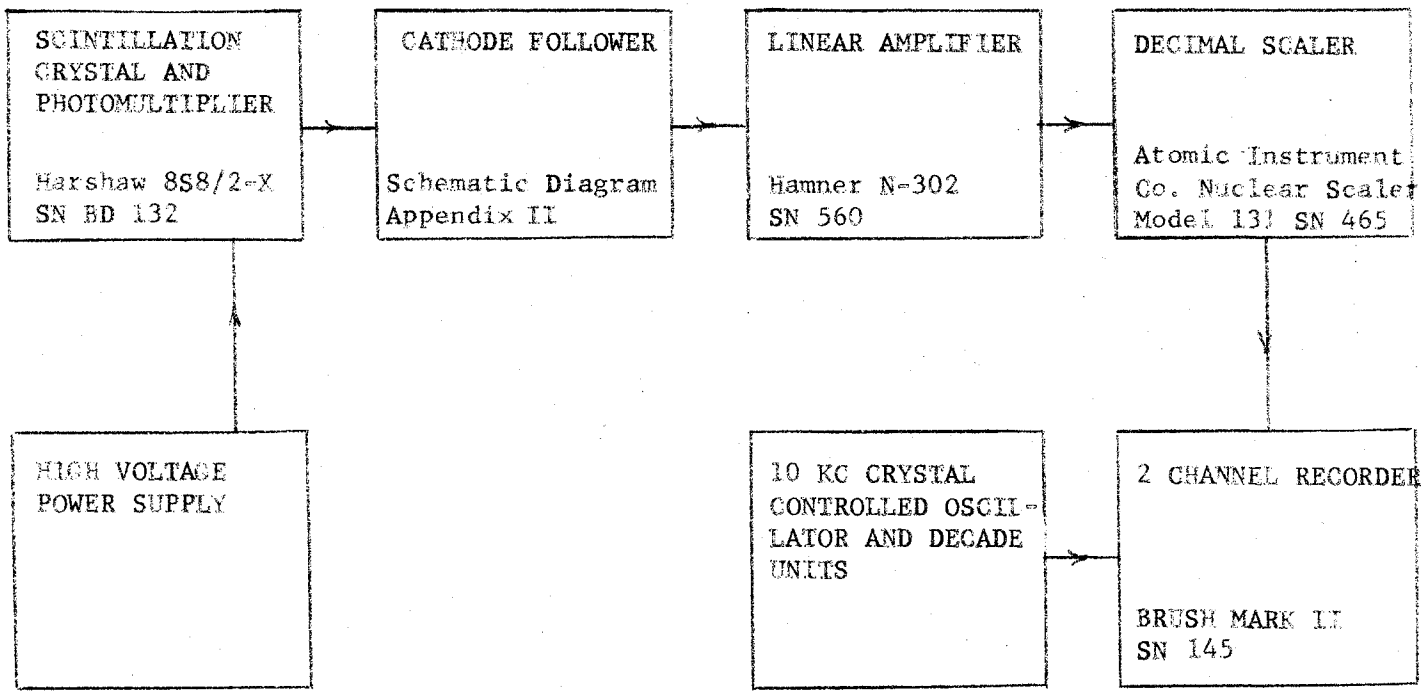

SCINTILLATION CRYSTAL AND PHOTOMULTIPLIER
Harshaw 8S8/2-X SN BD 132
CATHODE FOLLOWER
Schematic Diagram Appendix II
LINEAR AMPLIFIER
Hamner N-302 SN 560
DECIMAL SCALER
Atomic Instrument Co. Nuclear Scaler Model 131 SN 465
HIGH VOLTAGE POWER SUPPLY
10 KC CRYSTAL CONTROLLED OSCILLATOR AND DECADE UNITS
2 CHANNEL RECORDER
BRUSH MARK II SN 145

Figure 8. Alternate Counting System

tick mark on the recorder chart which was moving at a constant rate. The elapsed time between two marks was determined by measuring the separation on the chart paper. This time interval was compared with the time calibration made on the other channel. The equipment was assembled and tested, but has not actually been used to obtain data.

Figure 9 is a photograph of all of the assembled apparatus.

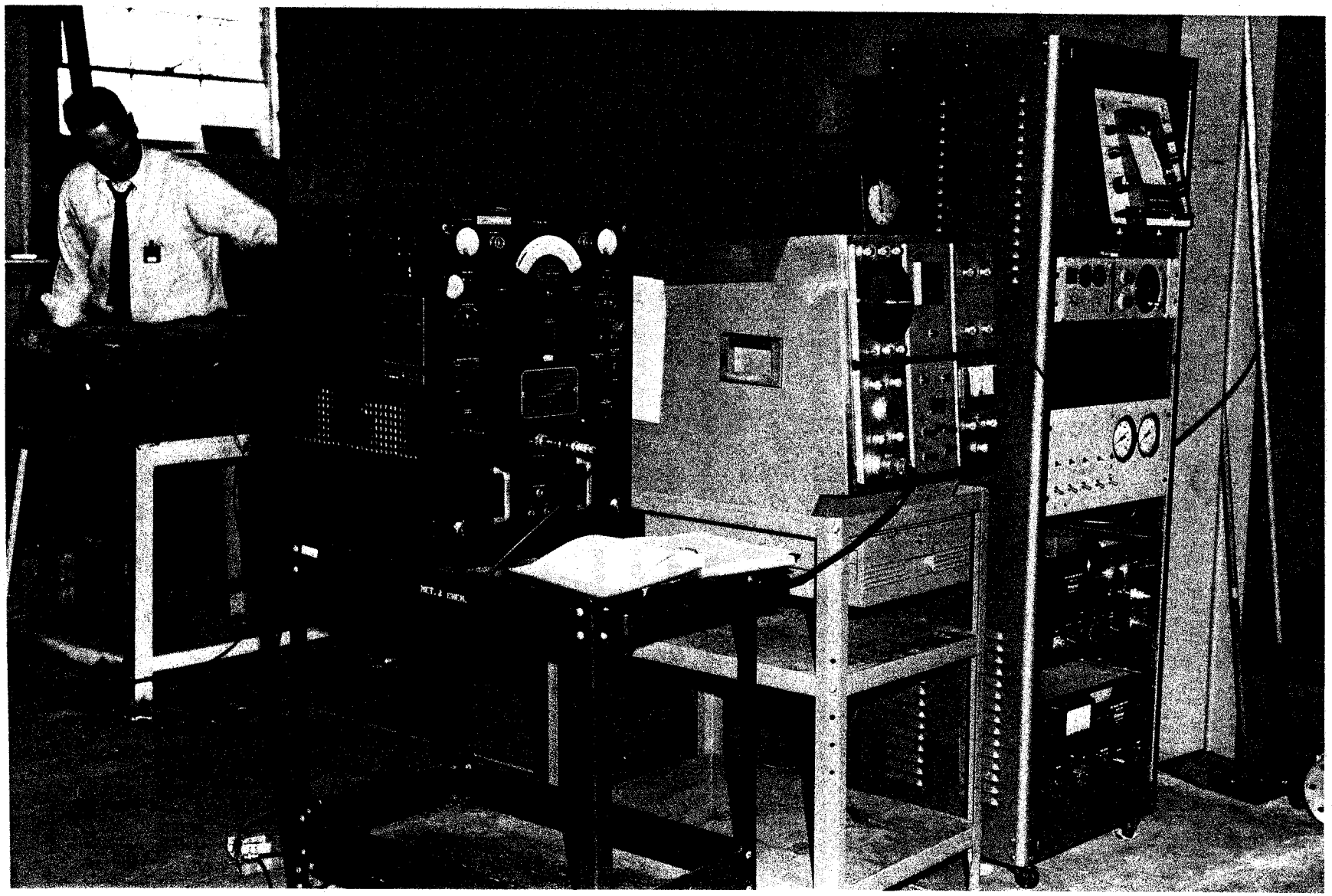


Figure 9. Counting Apparatus

IV. CALIBRATION

Three calibrations were required for the measurement of half-lives using the 128-channel analyzer; the live time and dead time calibration for the "gross mode" of operation, and the energy (pulse height) vs. channel number calibration for the "pulse height analyzer" mode of operation.

The live time is the time available for counting pulses in each of the channels. The live time is nearly equal to the time per channel, differing only by the constant time required for the analyzer to shift between channels. The time per channel, referred to as Δt, cannot be readily measured directly, and thus it is necessary to refer to the live time, which closely approximates Δt, for an estimate of the statistical variation of Δt. The calibration was accomplished by inserting pulses from a 10 KC crystal controlled oscillator into the second stage of the photomultiplier preamplifier. An analysis of the time calibrations made during an eight day period gave live times of .1902 $\pm$ .0006 seconds, .3804 $\pm$ .0007 seconds, and .7601 $\pm$ .0005 seconds for the .19 x 1, .19 x 2, and .19 x 4 seconds-per-channel time settings. The Δt's for the same settings are .192, .384, and .768 seconds per channel. The live time drift observed during the eight day period appeared to be within the above statistical variations of the live times. As an additional safeguard, checks of live times were made both before and after the taking of data.

Attempts to determine the dead time - the time following the incidence of a pulse that the counter will not detect a second pulse - were unsuccessful. Measurements were taken using the two source method, which gave dead times of 150, 190, and 170 microseconds for three successive trials for three different energy settings. This variation with energy is not anticipated from the design of the analyzer. The dead time measurements were made some time after the decay measurements, and it is suspected that instrument malfunction at the time of calibration is responsible for the poor results. If the actual instrument dead time is as large as 150 μsec, the maximum number of counts that can be recorded in a .19 second interval without excessive coincidence correction (defined for our purposes as 5%) is 300. This sets a limit of 6% statistical error on the counting rate determination by a single channel. During the calibration it was not possible to record sufficient counts using the .19 x 2 second channel time setting to permit statistically

sound measurements. It was noted that at approximately 800 counts per channel the subsequent positioning of the calibration sources closer to the scintillation crystal initially had no effect. Then, even closer positioning caused the count to drop to zero. This apparent saturation effect has not been reconciled with the counting of 10,000 counts per second during the live time calibration. Since it is anticipated that the alternate counting system will be used in the future, (see Recommendations) no further attempts were made to determine the precise dead time.

The energy versus pulse height calibration was made using the "pulse height analyzer" mode of operation of the 128-channel analyzer. Samples of Cs^{137}, Au^{198}, and Sm^{153} having known gamma energies of .6616 MEV for Cs, .4118 and .069 MEV for Au, and .1032 MEV for Sm were used to obtain the calibration curve (Figure 10). The curve is a plot of the channel number at which the maximum count occurs in the photopeak, versus the known energy of the photopeak.

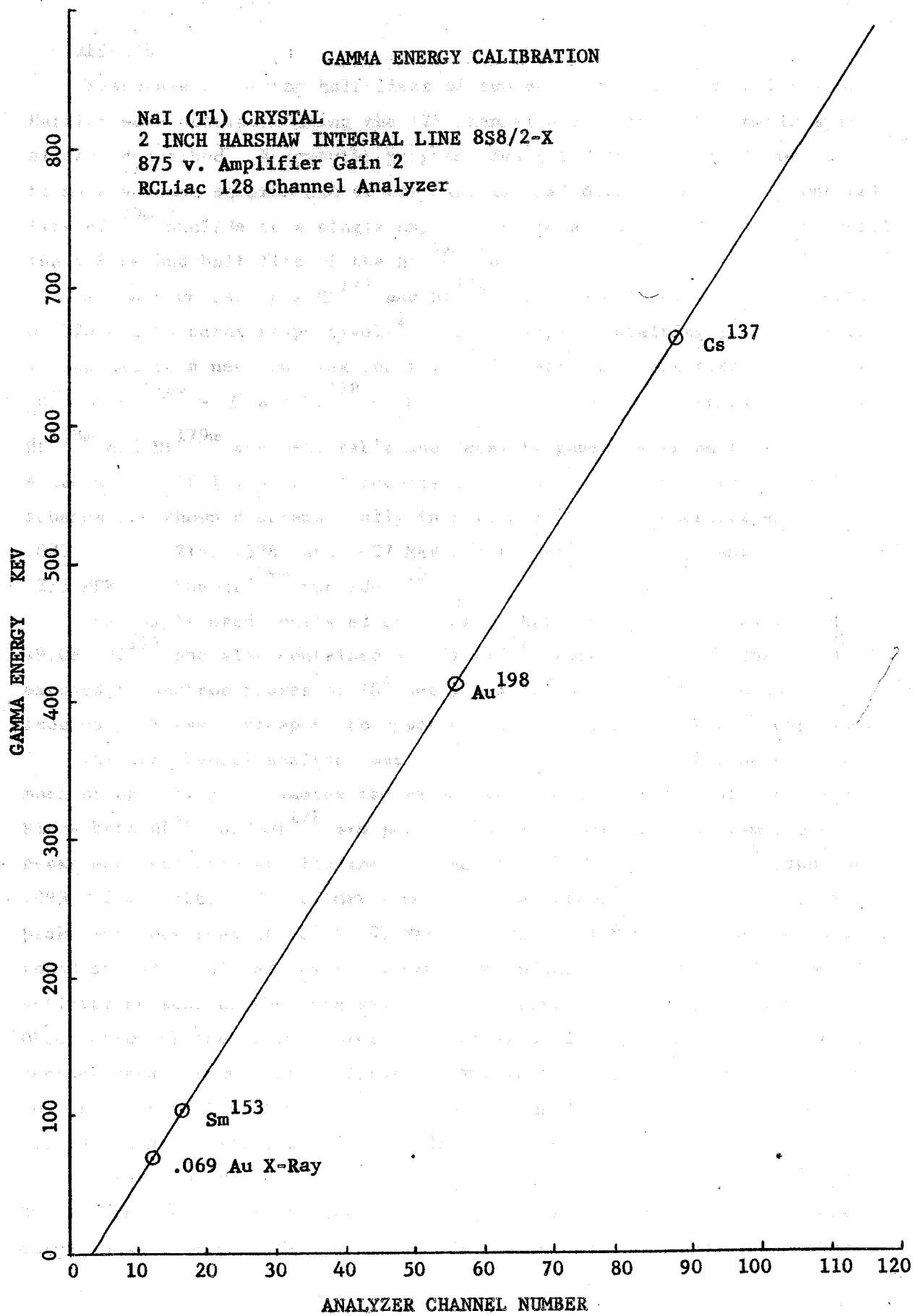


Figure 10. Gamma Energy Calibration

V. RESULTS

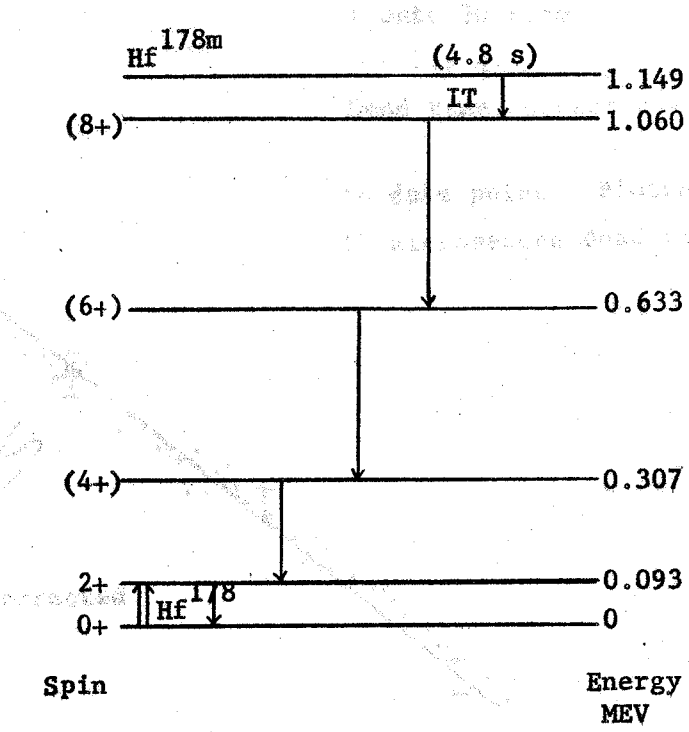
Measurements of the half-lives of two neutron activated nuclides of Hafnium were attempted using the 128-channel analyzer and pneumatic system already described. A computer program, described in Appendix I, was used to make a least squares fit of the experimental data for the 19 second half-life Hf^{179m} nuclide to a single exponential decay curve. Attempts to verify the 4.8 second half-life of the Hf^{178m} nuclide were unsuccessful.

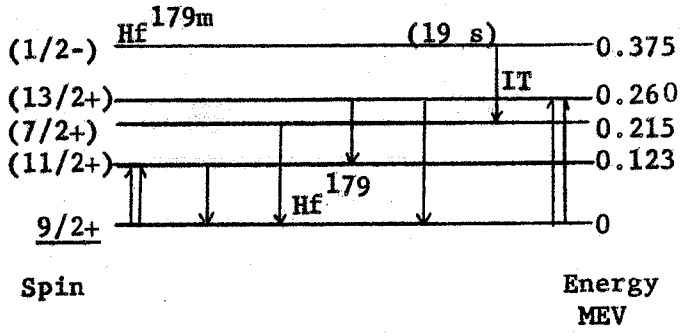
The Hafnium isotopes Hf^{177} and Hf^{178} have thermal neutron cross sections of 370 and 80 barns respectively.[2] When a sample containing these nuclides is exposed to a neutron flux the following reactions take place: $Hf^{177} + {}_0n^1 \longrightarrow Hf^{178m} + \gamma$ and $Hf^{178} + {}_0n^1 \longrightarrow Hf^{179m} + \gamma$. The resulting nuclides Hf^{178m} and Hf^{179m} are metastable and decay by gamma emission to the ground state with half lives of 4.8 seconds and 19 seconds respectively. The decay schemes are shown diagramatically in Figure 11. Gamma energies emitted are .089, .093, .214, .326, and .427 MEV for the Hf^{178m} nuclide and .160 and .215 MEV for the Hf^{179m} nuclide.[1,3]

The sample used consisted of 50 mg of Hafnium in oxide form enriched to 59.08% Hf^{177} and also contained 31.73% Hf^{178} (Appendix III). The sample was exposed to neutron fluxes of 10^8 and 2.5×10^9 neutrons/cm^2-second for 30 seconds. (These correspond to reactor powers of 2 and 50 watts respectively.)

The 128-channel analyzer was initially used in the pulse height analyzer mode of operation to examine the gamma spectrum of the activated Hafnium. Since both Hf^{177} and Hf^{178} are present in large percentages, gamma energy peaks were expected at .160 and .215 MEV from the decay of Hf^{179m} and .089, .093, .214, .326, and .427 MEV from the decay of Hf^{178m} (Figure 11). Major peaks were observed at .07 $\pm$ .01 MEV and .22 $\pm$ .01 MEV. Smaller peaks were found at .33 $\pm$ .01 and .44 $\pm$ .01 MEV. The peaks were attributed to gamma activity because of the beta shielding inherent in the detector arrangement. Observation of the sample immediately after activation, and then after 30 seconds decay, showed the .07 and .22 MEV peaks to be due primarily to the longer lived nuclide i.e., Hf^{179m}. The .22 MEV activity is attributed to the 19 second nuclide, and the .07 MEV corresponds to the Hafnium X-ray.

A window of approximately .04 MEV width was established at .22 $\pm$ .01 MEV and the gross mode of analyzer operation was used to obtain the decay curve shown in Figure 12. A Δt of .19 x 4 seconds per channel was chosen





DISINTEGRATION ENERGY AND SCHEME
Hf^{178m} and Hf^{179m}

Figure 11. Disintegration Energy and Scheme, Hf^{178m} and Hf^{179m}

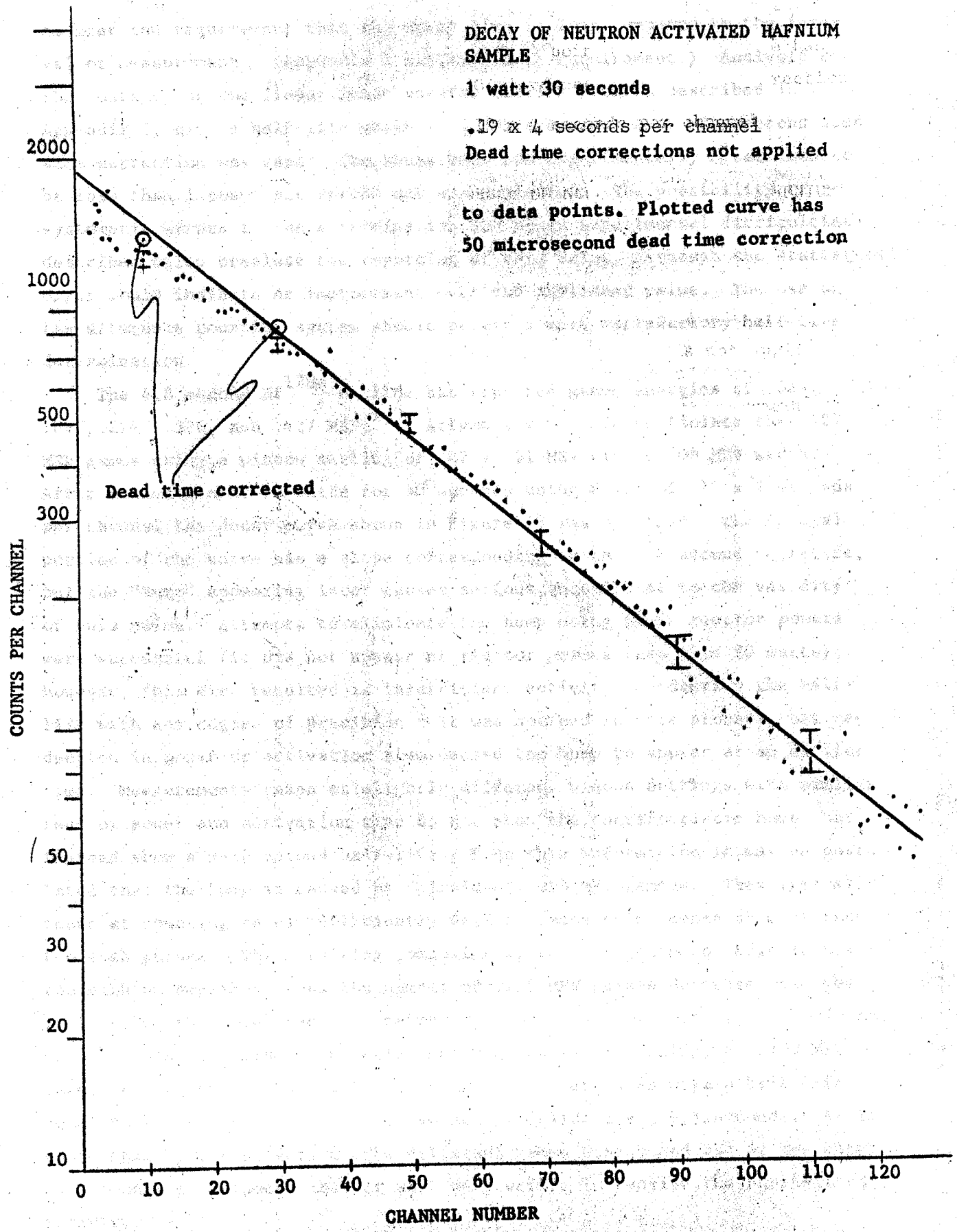


Figure 12. Decay of Neutron Activated Hf Sample

to meet the requirement that the decay time is long compared to the interval of measurement. (Appendix I explains this requirement.) Analysis of these data using the linear least squares fitting program described in Appendix I, gave a half-life of 18.15 $\pm$ .05 seconds. A 50 microsecond dead time correction was used. The background was experimentally determined to be less than 1 count per second and was neglected. The possibility of systematic errors in the determination and other experimental difficulties described below preclude the reporting of this value, although the statistical error would indicate an improvement over the published value. The use of the alternate counting system should permit a more satisfactory half-life determination.

The 4.8 second Hf^{178m} nuclide has reported gamma energies of .089, .093, .214, .326, and .427 MEV.[1,3] Attempts were made to isolate the .427 MEV gamma using a window setting of .43 $\pm$ .01 MEV with a .04 MEV width. After activation at 70 watts for 30 seconds using a Δt of .19 x 2 seconds per channel the decay curve shown in Figure 13 was obtained. The initial portion of the curve has a slope corresponding to the 4.8 second half-life, but the "hump" appearing later causes serious question as to the validity of this value. Attempts to eliminate the hump using lower reactor powers were successful (it did not appear at reactor powers less than 20 watts); however, this also resulted in insufficient activity to identify the half-life with any degree of precision. It was noticed in this process that reduction in power or activation time caused the hump to appear at an earlier time. Measurements taken at slightly different window settings with similar reactor power and activation time do not show the characteristic hump, but instead show a 9-10 second half-life. From this information it may be postulated that the hump is caused by coincident .215 MEV gammas. They initially occur at counting rates sufficiently high to cause coincidence of more than two such pulses. The resulting composite pulse is rejected because it has too high an amplitude. As the number of .215 MEV pulses decrease, the possibility of the coincidence of pairs of pulses, as opposed to the coincidence of more than two pulses, increases and thus pulses equivalent to .430 MEV gammas are counted. The number of such pulses decreases with a half-life equal to 1/2 the half-life of the decaying nuclide i.e., 9.5 seconds. It is hoped that by the selection of a different gamma energy and use of the alternate counting equipment that it will be possible to resolve the short-lived activity.

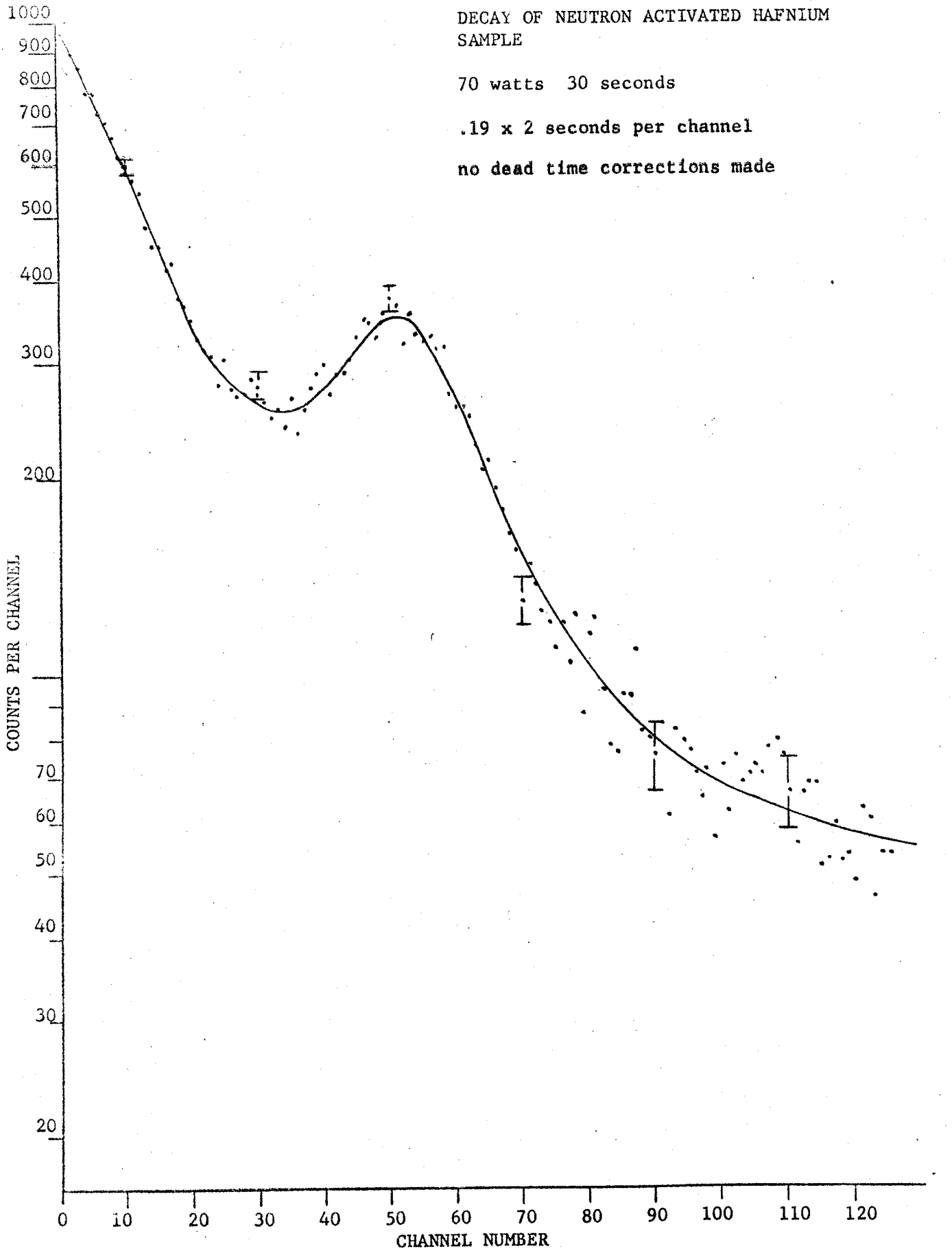


Figure 13. Decay of Neutron Activated Hf Sample

VI. RECOMMENDATIONS

Several modifications and additions to the present pneumatic tube system are recommended in order that the system be of maximum benefit to the U.S. Naval Postgraduate School in future work. These are outlined as follows:

1. Increase the vacuum reservoir tank from the present 314 cubic inch volume to approximately 1500 cubic inches. This will permit a faster evacuation of air from the tube prior to transfer of the rabbit into the core position. This allows a second rapid cycling of the rabbit if desired.

2. Install an automatic variable timing mechansim to replace the present manual switches. (The switches effectively determine pressure and vacuum in the tube by controlling the five solenoid valves.) This would make the cycling process a completely automatic procedure with the capability of changing irradiation and counting times as necessary.

3. Construct a special seating arrangement at the counting position of the pneumatic tube. This feature would permit the removal of the rabbit from the counting position without removing the seat from the pneumatic tube. In constructing this modification, thought should be given to the possibility of also measuring beta activity at the counting position.

The following recommendations are made concerning the measurements of short half-lives:

1. That the alternate system of counting (Figure 8) be used in lieu of the 128-channel analyzer. This would result in a reduction of both systematic errors and statistical errors. However, the 128-channel analyzer is very useful for exploratory decay measurements and measuring the energy spectrum of a particular sample.

2. That measurements of Hf continue using the alternate system of counting.

3. That measurements be made on Yb^{176} and Ba^{135}. 50 mg samples of Yb and Ba enriched in Yb^{176} and Ba^{135} respectively are presently available.

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APPENDIX I

Theory and Computer Program

Assume that the activity of a particular sample of a mixture of two independent nuclides having different half-lives and a constant background is given by the expression:

$$\dot{n} = \dot{n}_{10}e^{-\lambda_1 t} + n_{20}e^{-\lambda_2 t} + c \qquad (1)$$

where $\dot{n}_{10}$ and $\dot{n}_{20}$ are the initial activities at $t = 0$, λ_1 and λ_2 are the two decay constants and c is the constant background activity. It is desired to experimentally determine the five parameters appearing in equation (1), and in particular to determine the decay constants λ_1 and λ_2.

Experimentally one measures the total number of counts ΔN occurring in a known interval of time Δt. It is common practice to specify the decay time t to be at the midpoint of the interval Δt. Referring to Eq (1)

$$\Delta N = \int_{t-\frac{\Delta t}{2}}^{t+\frac{\Delta t}{2}} \dot{n}\, dt$$

and the experimentally determined mean counting rate over the interval Δt is

$$\dot{N} \equiv \frac{\Delta N}{\Delta t} = \frac{1}{\Delta t}\int_{t-\frac{\Delta t}{2}}^{t+\frac{\Delta t}{2}} \dot{n}\, dt$$

Using Eq (1)

$$\dot{N} = \frac{\dot{n}_{10}}{\lambda_1 \Delta t} e^{-\lambda_1 t}\left(e^{\frac{\lambda_1 \Delta t}{2}} - e^{-\frac{\lambda_1 \Delta t}{2}}\right) + \qquad (2)$$

$$\frac{\dot{n}_{20}}{\lambda_2 \Delta t} e^{-\lambda_2 t}\left(e^{\frac{\lambda_2 \Delta t}{2}} - e^{-\frac{\lambda_2 \Delta t}{2}}\right) + c$$

we now make the following assumptions about Δt and the decay constants:

$\lambda_1 > \lambda_2$ (Nuclide 1 decays faster than nuclide 2.)

$\Delta t < \frac{1}{\lambda_1}$ (The measuring interval is small compared to the mean life of nuclide 1, and thus even smaller than the mean life of nuclide 2.)

Expanding the exponential terms in a power series, and neglecting terms of smaller than second order results in the approximation to Eq. (2) of:

$$\dot{N} = \dot{n}_{10} e^{-\lambda_1 t}\left[1 + \frac{(\lambda_1 \Delta t)^2}{24}\right] + \dot{n}_{20} e^{-\lambda_2 t}\left[1 + \frac{(\lambda_2 \Delta t)^2}{24}\right] + C \quad (3)$$

If Δt is 1/10 of the mean life, then the correction factor (second order term) in the first bracket is $\sim 4 \times 10^{-4}$ and the corresponding correction for the second term is even smaller. Henceforward it will be assumed that Δt is always chosen small enough so that the above correction terms are negligible compared to the statistical uncertainty of the experimentally determined $\dot{N}$. Thus $\dot{N} = \dot{n}$ and the form of Eq. (1) can be used. If this were not so, Eq. (2) or Eq. (3) would have to be used for the experimental determination of λ_1 and λ_2.

Therefore $\dot{N} = n_{10} e^{-\lambda_1 t} + \dot{n}_{20} e^{-\lambda_2 t} + c$ (4)

where $\dot{N} = \frac{\Delta N}{\Delta t}$ and t is always specified to be at the midpoint of the constant time interval Δt.

If activity 2 and the background activity were completely missing then $\dot{N} = \dot{n}_{10} e^{-\lambda_1 t}$, and $\ell n\, \dot{N} = \ell n\, \dot{n}_{10} - \lambda_1 t$ is the resulting linear equation which can then easily yield a least square determination of λ_1.

If activity 2 is not present, but a background activity c is present, then

$\ell n\ (\dot{N}-c) = \ell n\ \dot{n}_{10} - \lambda_1 t$ can be least squares fitted to determine λ_1.

This procedure assumes that c is previously experimentally determined. If c is not independently determined then the best values of $\dot{n}_{10}$, λ_1, and c will be determined by a least squares fit to the equation:

$$\dot{N} = \dot{n}_{10} e^{-\lambda_1 t} + c.$$

A least squares fit of the observed counting rate $\dot{N}$ and t has been devised using a Taylor series approximation*. If

$\dot{N} = \dot{n}_{10} e^{-\lambda_1 t} + \dot{n}_{20} e^{-\lambda_2 t} + c$ is differentiated with respect to

$\dot{n}_{10}$, $\dot{n}_{20}$, λ_1, λ_2, and c,

then $\frac{\partial \dot{N}}{\partial \dot{n}_{10}} = e^{-\lambda_1 t}$, $\frac{\partial \dot{N}}{\partial \dot{n}_{20}} = e^{-\lambda_2 t}$,

$\frac{\partial \dot{N}}{\partial \lambda_1} = -\frac{\dot{n}_{10}}{t} e^{-\lambda_1 t}$, $\frac{\partial \dot{N}}{\partial \lambda_2} = -\frac{\dot{n}_{20}}{t} e^{-\lambda_2 t}$, and $\frac{\partial \dot{N}}{\partial c} = 1$.

The Taylor series expansion for a particular point $\dot{n}_i$, t_i using only the first derivatives becomes

$$\dot{n}_i = \bar{\dot{n}}_i + e^{-\bar{\lambda}_1 t_i} \Delta \dot{n}_{10} + e^{-\bar{\lambda}_2 t_i} \Delta \dot{n}_{20} - \frac{\bar{\dot{n}}_{20}}{t_i} e^{-\bar{\lambda}_2 t_i} \Delta \lambda_2 - \frac{\bar{\dot{n}}_{10}}{t_i} e^{-\bar{\lambda}_1 t_i} \Delta \lambda_1 + \Delta c \qquad (5)$$

where $\bar{\lambda}_1, \bar{\lambda}_2, \bar{\dot{n}}_{10}, \bar{\dot{n}}_{20}, \bar{\dot{n}}_i, \bar{c}$ represent approximations to $\lambda_1, \lambda_2, \dot{n}_{10}, \dot{n}_{20}, \dot{n}_i, c$

and $\Delta \dot{n}_{10} = \dot{n}_{10} - \bar{\dot{n}}_{10}$, $\Delta \dot{n}_{20} = \dot{n}_{20} - \bar{\dot{n}}_{20}$ $\Delta \lambda_1 = \lambda_1 - \bar{\lambda}_1$, $\Delta \lambda_2 = \lambda_2 - \bar{\lambda}_2$, $\Delta c = c - \bar{c}$.

The best values of $\dot{n}_{10}$, $\dot{n}_{20}$, λ_1, λ_2 and c are found first by writing i

*This method has been suggested by Prof. G.W. Rodeback who has applied it to least squares fitting of hyperbolic sine and other non-linear functions.

equations of the form:

$$\sqrt{\omega_i}\left\{e^{-\bar{\lambda}_1 t_i}\Delta\dot{n}_{10} + e^{-\bar{\lambda}_2 t_i}\Delta\dot{n}_{20} - \frac{\bar{\dot{n}}_{10}}{\bar{\tau}_1}e^{-\bar{\lambda}_1 t_i}\Delta\lambda_1 - \frac{\bar{\dot{n}}_{20}}{\bar{\tau}_2}e^{-\bar{\lambda}_2 t_i}\Delta\lambda_2 + \Delta c\right\} = \sqrt{\omega_i}\left(\dot{N}_i - \bar{\dot{n}}_i\right) \quad . \tag{6}$$

Then the corrections $\Delta\dot{n}_{10}$, $\Delta\dot{n}_{20}$, $\Delta\lambda_1$, $\Delta\lambda_2$, and Δc are found by matrix methods resulting in improved values for $\bar{\lambda}_1$, $\bar{\lambda}_2$, $\bar{\dot{n}}_{10}$, $\bar{\dot{n}}_{20}$, c. This process is repeated until the corrections become arbitrarily small. In each step the term

$$\sum\left[\omega_i\left(\dot{N}_i - \bar{\dot{n}}_i\right)^2\right]$$

is minimized. ω_i, the weighing factor, is inversely proportional to the square root of the number of counts corresponding to the ith interval since this represents the standard deviation of $\dot{N}_i$.

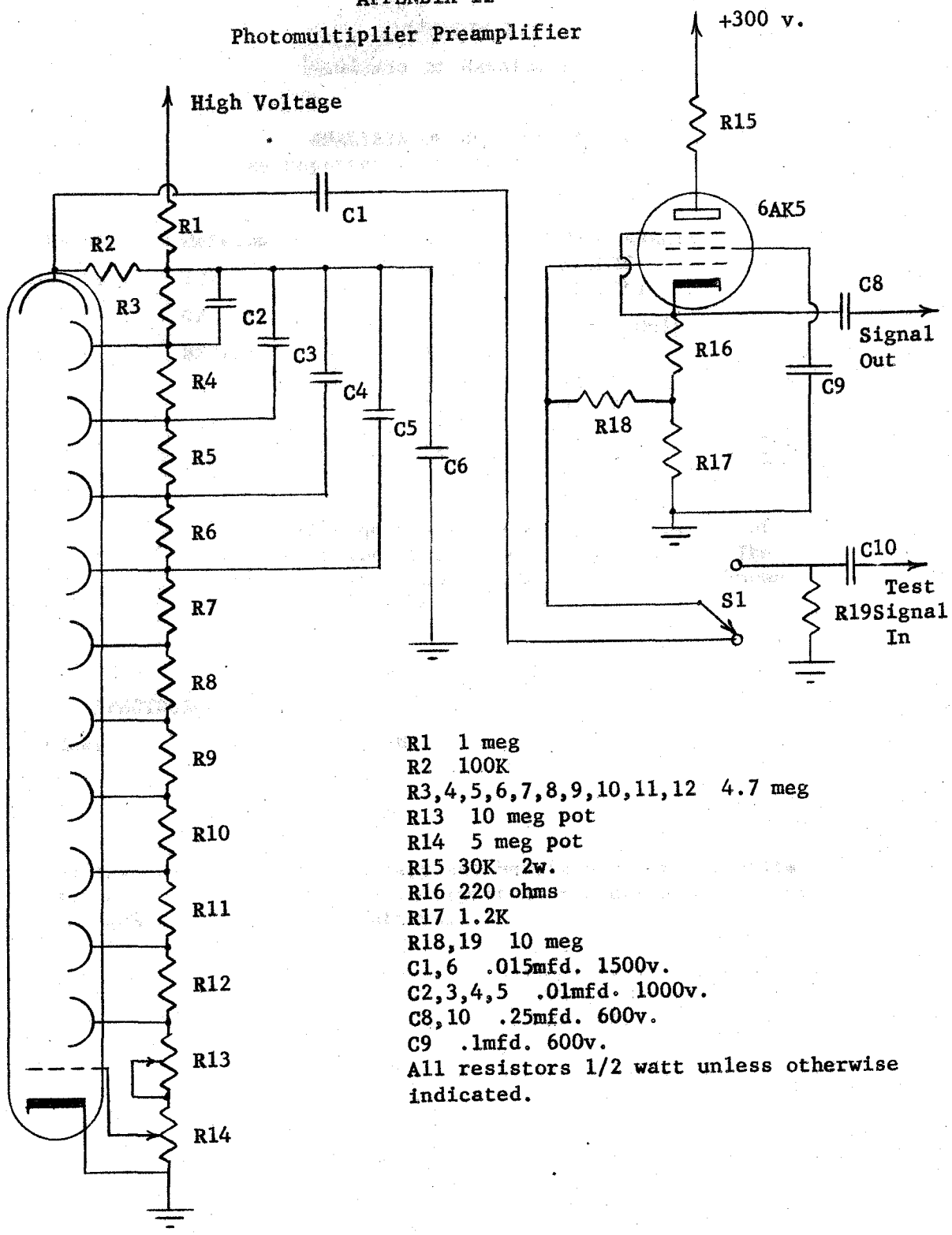
The program for the IBM 1604 Computer, as devised by personnel of the U.S. Naval Postgraduate School computer center, is attached. Results from trial runs using this method with both numerically devised data, and experimentally determined data for silver activated in the AGN 201 reactor, have been obtained. In the case of silver activities, the half lives were determined as 23.2 seconds and 2.3 minutes, which is in agreement with the reported[1] values of 24 seconds and 2.3 minutes for Ag^{110} and Ag^{108} respectively.

```
 WILLIAMS
  PROGRAM FLUX
0DIMENSION  X(150,5),PHIO(150),PHIC(150),T(150),DELPHI(150),W(150),
1H(5),XW(5,150),A(5,6),DELCO(5)
  READ 1, C1,CK1,C2,CK2,C3
  FORMAT(5F10.0)
  KK=0
  READ 2,N,DELT,L
  FORMAT(I3,F3.1,I3)
  READ 31 ,(T(I),PHIO(I),I=1,N)
  FORMAT(6(F4.2,3X,F5.0))
  DO 32 I =2,N
  T(I) = T(I) + T(I-1)
  DO 200 I=1,N
  CHANL = L+I-1
  CNST=38.E-6 +0.5E-6*CHANL
  PHIO(I)=PHIO(I)/(1.-PHIO(I)*CNST/DELT)
  DO 100 I = 1,N
  W(I) = SQRTF(PHIO(I))
  DO 10 I=1,N
  TI=T(I)
  EK1T = EXPF(CK1*TI)
  EK2T = EXPF(CK2*TI)
  PHIC(I)=C1*EK1T+C2*EK2T +C3
  X(I,1)=EK1T
  X(I,3) = EK2T
  X(I,2) = C1*TI*EK1T
  X(I,4) = C2*TI*EK2T
  X(I,5)=1.0
  DELPHI(I) = PHIO(I) - PHIC(I)
  PRINT 8 (DELPHI(I) , I = 1,N)
  DO 11 J=1, N
  W=W(J)
  DO 11 I= 1,5
  XW(I,J) = W*X(J,I)
  DO 13 I=1,5
  H(I)=0.
  DO13 K=1,N
  H(I)=H(I) +XW(I,K)*DELPHI(K)
  DO 15 I = 1,5
  DO 15 J = 1,5
  P=0.
  DO 14 K=1,N
  P=P+XW(I,K)*X(K,J)
  A(I,J)=P
  DO 12 I=1,5
  A(I,6) = H(I)
  PRINT 300
  FORMAT(4H DEL)
  CALL JORDAN2(A,5,DELCO)
  DNORM = 0.
  DO 16 I=1,N
  DNORM = DNORM + DELPHI(I)*DELPHI (I)
  EST=0.
  DO 17 I=1,5
  EST=EST + DELCO(I)*H(I)
  PRINT 301 , DNORM,EST
  FORMAT(2E12.5)
  PRINT 7 KK,C1,CK1,C2,CK2,C3
  FORMAT(2X,I2,5X,5E15.6)
  PRINT 8 (DELCO(I),I=1,5)
```

```
8 FORMAT(7X, 5E17.7)
  C1 = C1+DELCO(1)
  CK1= CK1 +DELCO(2)
  C2 = C2 + DELCO(3)
  CK2 = CK2 + DELCO(4)
  C3 = C3 + DELCO(5)
  KK = KK+1
  IF(KK -20) 20,20,30
0 PRINT 40
0 FORMAT(5H1JOB2)
  GO TO 33
  END
  SUBROUTINE JORDAN2(A,N,X)
  DIMENSION A(5,6) ; X(5)
  K =N+1
1 IF (K-1)13,6,15
5 D=0.
  DO 2 I=2,K
  IF (ABSF(A(I-1,1))  -D) 4,4,3
3 L =I-1
2 D =ABSF(A(L,1))
4 IF(L-1)5,6,5
5 DO 7 J=1,K
  D =A(L,J)
  A(L,J) = A(1,J)
7 A(1,J) = D
6 DO 8 I =1,N
8 X(I) = A(I,1)
  IF (K-1)12,13,12
2 DO 10 J=2,K
  D = A(1,J)/X(1)
  DO   9 I=2,N
9 A(I-1,J-1)  = A(I,J) -X(I)*D
0 A(N,J-1) =D
  K = K-1
  GO TO 11
3 CONTINUE
  RETURN
  END
  END
```

APPENDIX II

Photomultiplier Preamplifier



R1 1 meg
R2 100K
R3,4,5,6,7,8,9,10,11,12 4.7 meg
R13 10 meg pot
R14 5 meg pot
R15 30K 2w.
R16 220 ohms
R17 1.2K
R18,19 10 meg
C1,6 .015mfd. 1500v.
C2,3,4,5 .01mfd. 1000v.
C8,10 .25mfd. 600v.
C9 .1mfd. 600v.
All resistors 1/2 watt unless otherwise indicated.

APPENDIX III

Analysis of Hafnium Sample

ANALYSIS OF HAFNIUM SAMPLE
(as reported by Oak Ridge analysis report)

ELEMENT	Hafnium
ISOTOPE	177
SERIES	GP
SAMPLE	827(a)

ISOTOPIC ANALYSIS

ISOTOPE	ATOMIC PERCENT	PRECISION
174	Trace Only	
176	0.99	± 0.07
177	59.08	± 0.52
178	31.73	± 0.49
179	4.53	± 0.17
180	3.68	± 0.11

The limits quoted above are an expression of the precision of this measurement only. The error is estimated at less than 1% from known sources of systematic errors.

SPECTROSCOPIC ANALYSIS

ELEMENT	PERCENT
Cu	0.1
Fe	0.2

The spectrographic results reported herein are semi-quantitative estimates and should not be interpreted or construed to be precise quantitative determinations.